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Publication Date

2003-07-01

A CONCEPTUAL AND NUMERICAL MODEL FOR THERMAL-HYDROLOGICAL-CHEMICAL PROCESSES IN THE YUCCA MOUNTAIN DRIFT SCALE TEST

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Abstract: A numerical model was developed to predict the coupled thermal, hydrological, and chemical (THC) processes accompanying the Drift Scale Test (DST) at Yucca Mountain, NV. The DST has been closely monitored through the collection of gas, water, and mineral samples as well as thermal, hydrological, and mechanical measurements. A two-dimensional dual permeability model was developed to evaluate multiphase, multicomponent, reaction-transport processes in the fractured tuff. Comparisons between results using the TOUGHREACT code and measured water (e.g., pH, $SiO_2(aq)$, Na^+, K^+) and gas (CO_2) compositions show that the model captures the chemical evolution in the DST. Non-reactive aqueous species (e.g., Cl) show strong dilution in fracture waters, indicating little fracture-matrix interaction. Silica concentrations are higher than in the initial pore water and show a trend of increasing reaction with fracture-lining silicates at higher temperatures. The narrow precipitation zone of predominantly amorphous silica observed above the heaters was also captured.

1. INTRODUCTION

The long-term performance of the repository at Yucca Mountain will be affected by the coupling of thermal, hydrological and chemical (THC) processes in the rock around the emplacement drifts. The transport of heat, fluid, and vapor will result in changes in water and gas chemistry, as well as mineral dissolution and precipitation which may lead to permanent changes in porosity, permeability and unsaturated hydrological properties. The purpose of this contribution is to describe the approach used to model reaction-transport processes in the Drift Scale Test (DST) with comparisons of simulation results to measured geochemical data on water, gas, and minerals.

The DST is the second underground thermal test to be carried out in the Exploratory Studies Facility (ESF) at Yucca Mountain, Nevada. The purpose of the test is to evaluate the effect of coupled thermal, hydrological, chemical, and mechanical processes that take place in unsaturated fractured tuff over a range of temperatures (approximately 25°C to 200°C). The DST heaters were activated on December 3, 1997, with a planned period of 4 years of heating, followed by 4 years of cooling. After just over 4 years, the heaters were switched off and since that time the system has been slowly cooling.

The DST consists of an approximately 50 m long drift that is 5 m in diameter. Nine electrical floor canister heaters were placed in this drift (the Heated Drift) to simulate nuclear-waste-bearing containers. Electrical heaters were also placed in a series of horizontal boreholes (wing heaters) drilled perpendicular outward from the central axis of the Heated Drift. These heaters were emplaced to simulate the effect of adjacent emplacement drifts. Details regarding the DST layout, borehole orientations, operation of the test, and measurements performed are discussed in BSC (2002), CRWMS M&O (1998), and in Datta et al. (2003, this volume).

The major driving forces for THC processes in the thermal tests and in the potential repository are the thermal heat load and (over long time periods) boundary conditions such as infiltration rate. For the short time scale of the DST the infiltration rate is negligible, being on the order of 1 to 10 mm/year (Sonnenthal and Bodvarsson, 1999).

A recurring theme in this work is the importance of the interaction of flow and transport between fractures and the adjoining porous matrix. We note here that large-scale chemical equilibrium cannot be maintained in a system where mineral precipitation and dissolution are taking place at different rates in fractures and matrix (which have different mineral assemblages and abundances), the advective transport in fractures is faster than diffusive transport in the matrix.

The development of a conceptual model and the strategy of incorporating the important THC processes for the thermal tests are based originally on studies done on TH processes for the Single Heater Test and the DST (e.g., Tsang and Birkholzer, 1999; Birkholzer and Tsang, 1998) and a predictive model for THC processes in the DST (Sonnenthal et al., 1998; Xu et al., 2001). The following sections discuss some of the important aspects of the THC model, followed by comparisons to measured geochemical data.

2. DRIFT SCALE TEST THC MODEL

2.1 Conceptual model for THC processes

The evolution of the chemical regime in the unsaturated zone surrounding a heat source is closely related to the spatial distribution of temperature and the transport of water and vapor. An important aspect of the unsaturated fractured tuff at Yucca Mountain is that the highly permeable fractures are nearly dry, whereas the low permeability and porosity rock matrix has a water saturation of about 90 percent. Thus, heating of the rock induces boiling of the matrix pore water, transport of water vapor into fractures, and condensation along fracture walls.

The numerical model for reaction-transport processes in the fractured welded tuffs must account for the different rates of transport in fractures, compared to a much less permeable rock matrix. Transport rates greater than the rate of equilibration via diffusion leads to disequilibrium between waters in fractures and matrix. Because the system is unsaturated, and undergoes boiling, the transport of gaseous species, especially CO_2, is an important consideration. The model must also capture the differences in initial mineralogy in fractures and matrix and their evolution.

Although mechanical processes were certainly active during the DST, measured changes in air permeability were typically less than one order of magnitude (BSC, 2002). Permeability reductions could also be attributed to increases in fracture liquid saturation in the condensation zones. In addition, variations in fracture permeability are at least a few orders of magnitude around the DST and therefore smaller mechanical effects would not have a major effect on changes in water and gas chemistry resulting from coupled THC processes.

2.2 Numerical grid and properties

The two-dimensional dual-permeability grid for the DST represents a vertical cross section through the Heated Drift at a distance approximately 30 m from the bulkhead, separating the Heated Drift from the Access Drift (Figure 1; adapted from Birkholzer and Tsang, 1998).

The top boundary is approximately 99 m above the drift center, with the bottom boundary located approximately 157 m below the center. Small grid blocks are employed adjacent to the wing heaters and drift wall to capture the strong gradients in temperature and liquid saturation in these regions.

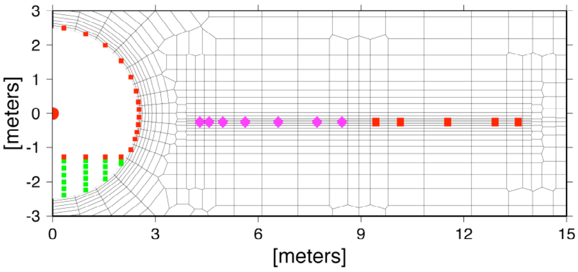

Figure 1. Partial view of 2-D numerical mesh for the DST (adapted from Birkholzer and Tsang, 1998). Symbols represent the central canister heater (red circle), wing heater locations (purple diamonds and red squares), concrete invert (green squares), and drift wall and invert top blocks (small red squares).

The Heated Drift grid block is connected directly to the Heater Test Alcove grid block and the connection area and distance were adjusted so that heat loss from the drift resulted in roughly similar crown temperatures to the maximum observed values. This was done to simulate heat and mass losses through the bulkhead, instead of reducing power by a factor. In the approximate location of the Observation Drift, the grid block volumes were increased to represent connection to the atmosphere. Details regarding the hydrological and thermal properties; thermodynamic and kinetic data, and; time-dependent heat input are discussed in Spycher et al. (2003).

2.3 Geochemical model

In order to handle separate yet interacting processes in fractures and matrix, the dual permeability method has been adopted, such that each grid block is divided into matrix and fracture continua, characterized by their own pressure, temperature, liquid saturation, water and gas chemistry, and mineralogy. Simulations of THC processes include coupling between heat, water, and vapor flow; aqueous and gaseous species transport; kinetic and equilibrium mineral-water reactions; and feedback of mineral precipitation/dissolution on porosity, permeability, and capillary pressure.

Aqueous species included in the model are H^+, Ca^{2+}, Na^+, K^+, $SiO_2(aq)$, Mg^{2+}, Al^{3+}, Fe^{3+}, SO_4^{2-},

HCO_3^-, Cl^-, F^-, and NO_3^-. The initial pore water chemistry is based on a sample that was ultracentrifuged from the proposed repository host rock (Tptpmn). Minerals considered include silica phases (□-cristobalite, quartz, tridymite, amorphous silica, and opal-CT), calcite, feldspars, smectites, illite, kaolinite, sepiolite, zeolites, fluorite, hematite, and gypsum. Treatment of CO_2 includes gas-water equilibration, diffusion, and advection.

2.4 TOUGHREACT code

Simulations of THC processes were performed using the TOUGHREACT code (Xu and Pruess, 2001; Xu et al., 2003). TOUGHREACT is based on TOUGH2 (Pruess, 1991) and in addition to the coupling between heat, water, and vapor flow for multiphase systems, it solves aqueous and gaseous species transport, kinetic and equilibrium mineral-water reactions, and feedback of mineral precipitation/dissolution on porosity, permeability, and capillary pressure.

TOUGH2 uses the integral finite difference method, with the equations for the multiphase system solved simultaneously, primarily with an iterative solver. Reaction-transport equations are solved using the sequential non-iterative operator splitting method, whereby each aqueous and gaseous component is transported separately by diffusion and advection, followed by solving the local kinetic and equilibrium mineral-water-gas reactions at grid block using Newton-Raphson iteration.

The rate of flow between fluids in fractures and in the rock matrix is a crucial factor for transport and reaction in fractures. For consistency with the formulation for flow between fractures and matrix used in the Yucca Mountain Project, the reactive surface area for minerals in unsaturated fractures has been related to the fracture-matrix interaction area based on a modified form of the "Active Fracture Model" (Liu et al., 1998; Sonnenthal et al., 2003). In this way, the wetted surface area for mineral-water reactions is consistent with that for flow and diffusion.

3. MODEL RESULTS AND COMPARISONS TO DATA

3.1 Gas-phase CO_2 evolution

The concentration of CO_2 in the gas phase is a function of temperature, pressure, aqueous-phase chemistry, mineral-water reactions, and advective and diffusive transport. From a model validation standpoint, the strong effect of CO_2 partial pressure on water pH and the final brine composition formed upon evaporation highlight the importance to the analysis of CO_2 concentrations. Simulation results are compared to concentrations measured in gas samples taken from boreholes during the heating phase of the DST. CO_2 concentrations in gases collected from the DST also provide a qualitative measure of the influence of atmospheric gas on the system, because of the relatively low and constant value in the atmosphere (□400 ppmv).

The modeled evolution of CO_2 has been validated by comparison to over 4 years of measurements from the DST. Simulated CO_2 concentrations in the fracture gas after 3 years of heating (Figure 2) show the general outward migration of elevated CO_2 concentrations with the boiling front. The peak in CO_2 concentrations takes place at approximately 60°C, and at higher temperatures the concentrations generally decline as a result of degassing and transport with water vapor to cooler regions.

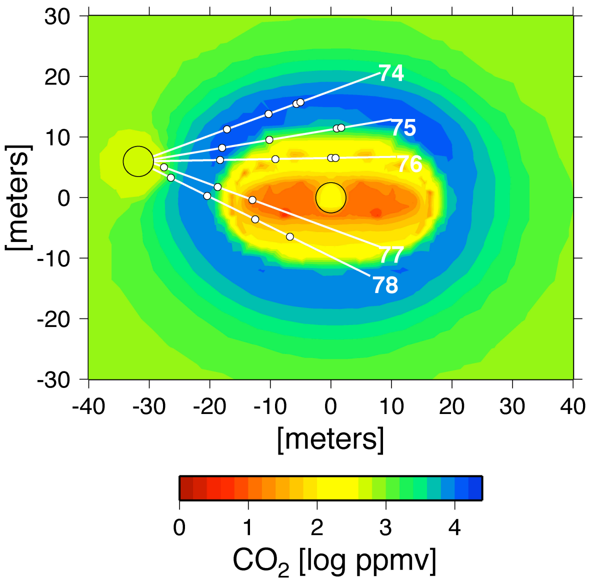

Figure 2. Modeled CO_2 concentrations in fractures after 3 years of heating. Note locations of numbered boreholes collared in the Observation Drift (circular region at left).

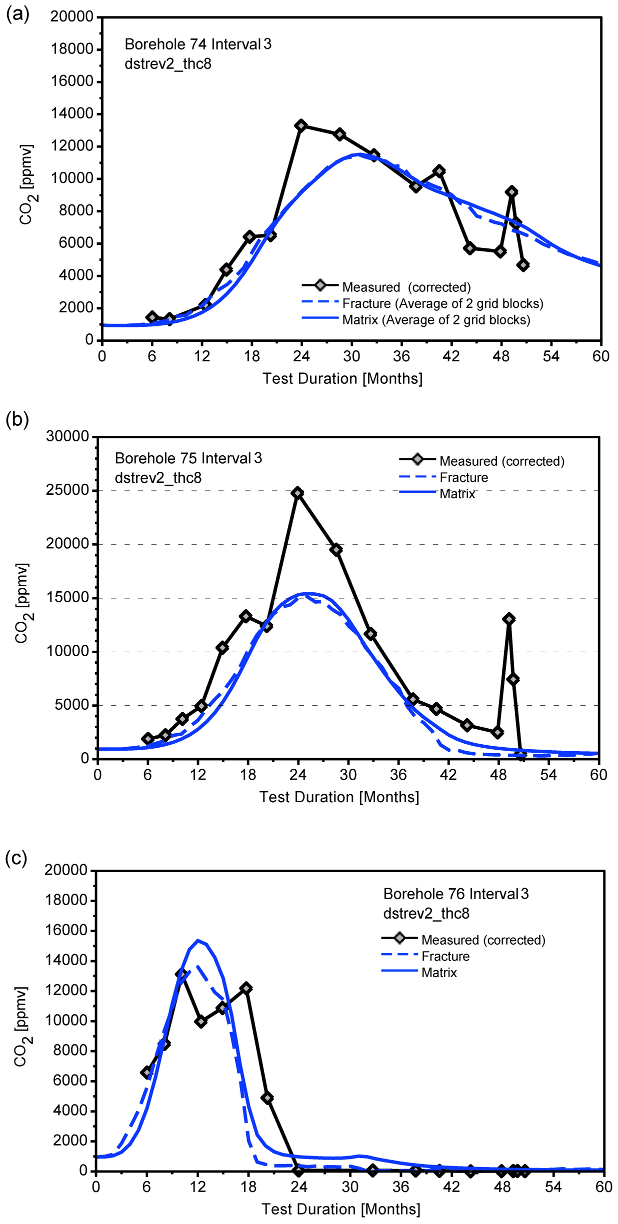

Figure 3. Modeled CO_2 concentrations in fractures and matrix compared to measured values from boreholes (corrected for vapor condensation) (a) Borehole interval 74-3 (average of bounding grid blocks); (b) Borehole interval 75-3; (c) Borehole interval 76-3.

Comparisons of modeled CO_2 to measurements performed on gas samples from boreholes (shown in Figure 2) are presented in Figure 3 (above). Samples were collected from zones a few meters (borehole interval 76-3) to about 15 meters away from the Heated Drift (borehole interval 74-3). Measured concentrations were corrected for water vapor condensation that took place as part of the procedure for gas sampling. Zones closest to the heaters (interval 76-3) exhibit narrower and earlier peaks in concentration compared to zones further out in the rock (interval 74-3).

Simulated and measured concentrations are close in magnitude and in their trends. There is little difference between fracture and matrix concentrations, because of rapid equilibration by advection and diffusion of gas species and their local equilibration with pore water.

3.2 Aqueous species evolution

The evolution of aqueous species in waters collected in the DST exhibit small reductions in pH (from about pH 8 in the pore water to about 6-8 in condensate waters. The drop in pH is related to the local increases in CO_2 concentrations. Figure 4 shows an example of the initial drop in pH during vapor condensation, followed by increasing pH as the zone is further heated and CO_2 is diluted by water vapor.

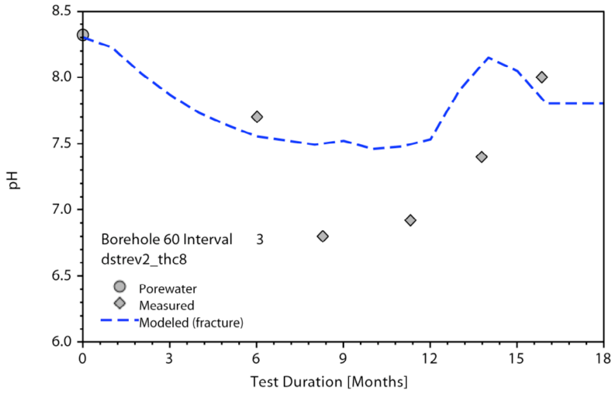

Figure 4. Measured and modeled pH (in fractures) for samples collected from borehole interval 60-3, located below the heaters.

Simulated and measured concentrations of conservative species in the fractures, such as chloride, are much lower than in the initial matrix pore water, indicating that fracture-matrix interaction has been negligible. However, reactive species, such as silica and potassium show significant effects of reaction with fracture-lining silicate minerals.

3.3 Mineral precipitation/dissolution

Model predictions, followed by analyses of *in-situ* sidewall core samples, showed that amorphous silica, calcite and lesser amounts of gypsum are the dominant phases expected to precipitate in the boiling regions. The greatest amount of mineral precipitation is predicted to be above the heaters where reflux of water condensed in fractures dissolves fracture-lining minerals and is boiled.

Simulations and measurements of amorphous silica and calcite, are shown in Figures 5 and 6. Amorphous silica forms only where strong evaporation by boiling takes place. In comparison, calcite that is originally present in fractures, dissolves in the lower pH waters in condensation zones around the boiling zone and in the drainage zones below the heaters. It also forms with amorphous silica and gypsum in the boiling zones.

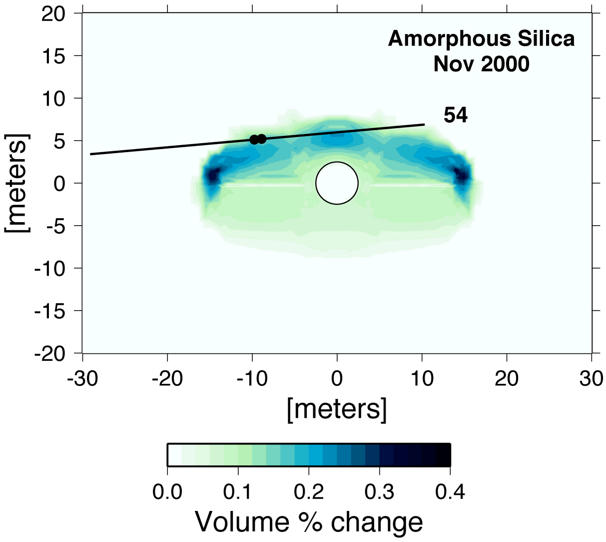

Figure 5. Volume percent change in amorphous silica abundance in fractures. Filled circle indicates sidewall core sample locations where it was observed.

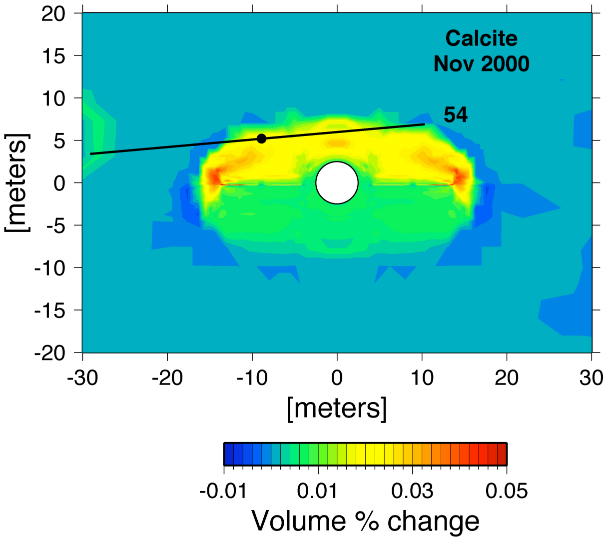

Figure 6. Volume percent change in calcite abundance in fractures. Filled circle indicates sidewall core sample locations where it was observed.

4. CONCLUSIONS

Prior to this study, little work had been done to model THC processes in unsaturated fractured rock under boiling conditions. The DST presented an unprecedented opportunity to test the conceptual models and codes on a well-constrained system, over time and spatial scales greater than simple lab experiments.

Comparisons of modeled concentrations and the chemistry of water (e.g., pH, SiO_2(aq), Na^+, K^+) and gas samples (CO_2) collected over 5 years show that the THC model captures the trends and magnitude of chemical changes. CO_2 concentrations in gas samples increase from an initial value of about 1000 ppmv to greater than 25,000 ppmv as a result of exsolution from matrix pore water and transport into fractures. The CO_2 dissolves into condensate waters at lower temperatures, resulting in a drop in pH and also dissolution of calcite.

Based on the strong dilution trends of non-reactive species, such as chloride and sulfate, the extent of interaction of fracture condensate water with matrix pore water must have been very limited. However, reactive species, such as silica, show increasing concentrations owing to reaction with predominantly fracture-lining silica polymorphs and feldspars at higher temperatures. A precipitation zone of secondary mineral phases such as amorphous silica, calcite, and gypsum in fractures has also been successfully predicted in a narrow reflux zone above the heaters. Consistent with the observation of coatings of amorphous silica that are tens of microns or less in thickness, the predicted fracture porosity change is less than one percent.

Because the chemical changes are so closely tied to the TH processes and fracture-matrix interaction, modeling several chemical components provides an independent method for validating and refining TH models (e.g., the "Active Fracture Model" for fracture-matrix interaction and the dual-permeability method). The methodology developed for the THC model of the DST can then be applied with more certainty to long-term predictions of nuclear waste repository performance.

5. ACKNOWLEDGEMENTS

Aqueous geochemical data from the Drift Scale Test were obtained by Laura DeLoach (Lawrence Livermore National Lab). Schon Levy (Los Alamos National Lab) determined the mineral abundances in fractures. Sumit Mukhopadhyay and Jens Birkholzer developed the initial TH grid from which the THC model was developed. Sonia Salah worked on the thermodynamic and kinetic databases. This work was supported by the Director, Office of Civilian Radioactive Waste Management, U.S. Department of Energy, through Memorandum Purchase Order EA9013MC5X between Bechtel SAIC Company, LLC, and the Ernest Orlando Lawrence Berkeley National Laboratory (Berkeley Lab). The support is provided to Berkeley Lab through the U.S. Department of Energy Contract No. DE-AC03-76SF00098.

6. REFERENCES

BSC (Bechtel SAIC Company) 2002. *Thermal Testing Measurements Report.* Las Vegas, Nevada: Bechtel SAIC Company.

CRWMS M&O (Civilian Radioactive Waste Management System Management and Operating Contractor) 1998. *Drift Scale Test As-Built Report.* Las Vegas, Nevada: CRWMS M&O.

Birkholzer, J. T. & Tsang, Y. W. 1998. *Interpretive Analysis of the Thermo-Hydrological Processes of the Drift Scale Test.* In: Drift Scale Test Progress Report, Chapter 6. Yucca Mountain Project Level 4 Milestone SP2930M4. Berkeley, California: Lawrence Berkeley National Laboratory.

Datta, R., Barr, D., & Boyle, W. 2003. *Measuring thermal, hydrological, mechanical, and chemical responses in the Yucca Mountain Drift Scale Test.* Proceedings (to be published), GeoProc2003, Stockholm, Sweden, October 2003.

Liu, H.H., Doughty, C., & Bodvarsson, G.S. 1998. *An active fracture model for unsaturated flow and transport in fractured rocks.* Water Resources Research, 34(10), 2633–2646.

Pruess, K. 1991. TOUGH2: *A general purpose numerical simulator for multiphase fluid and heat flow.* Lawrence Berkeley National Laboratory Report LBL-29400, Berkeley, California.

Sonnenthal, E.L., Bodvarsson, G.S. 1999. *Constraints on the hydrology of the unsaturated zone at Yucca Mountain, NV from three-dimensional models of chloride and strontium geochemistry.* J. Contam. Hydrol., 38, 107-156.

Sonnenthal, E., Spycher, N., Apps, J., & Simmons, A. 1998. *Thermo-Hydro-Chemical Predictive Analysis for the Drift-Scale Heater Test.* Yucca Mountain Project Level 4 Milestone SPY289M4. Berkeley, California: Lawrence Berkeley National Laboratory.

Spycher, N., Sonnenthal, E., Dobson, P., Apps, J., Salah, S., Kneafsey, T & Haukwa C. 2003. *Drift-Scale Coupled Processes (DST and THC Seepage) Models.* MDL-NBS-HS-000001 REV02. BSC: Las Vegas, NV.

Tsang, Y., Birkholzer, J. 1999. *Predictions and observations of the thermal-hydrological conditions of the Single Heater Test.* Journal of Contaminant Hydrology, 38: pp. 385-425.

Xu, T., & Pruess, K. 2001. *Modeling multiphase non-isothermal fluid flow and reactive geochemical transport in variably saturated fractured rocks: 1. Methodology.* American Journal of Science, 301: pp. 16-33.

Xu T., Sonnenthal, E., Spycher, N., Pruess, K., Brimhall, G., & Apps, J., 2001. *Modeling multiphase non-isothermal fluid flow and reactive geochemical transport in variably saturated fractured rocks: 2. Applications to supergene copper enrichment and hydrothermal flows.* American Journal of Science, 301: pp. 34-59.

Xu, T., Sonnenthal, E., Spycher, N., & Pruess, K. 2003. *TOUGHREACT: A new code of the TOUGH family of non-isothermal multiphase reactive geochemical transport in variably saturated media.* Proceedings, TOUGH Symposium, 2003.